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Influence of electron capture and loss in the energy distribution of protons dissociated from fast H_2^+ molecules in carbon foils

Isabel Abril[*,1], **Cristian D. Denton**[2], **Manuel D. Barriga-Carrasco**[3], **Rafael Garcia-Molina**[3], **Kenji Kimura**[4], and **Néstor R. Arista**[5]

[1] Departament de Física Aplicada, Universitat d'Alacant, Apartat 99, 03080 Alacant, Spain
[2] Departamento de Física, Universidad Técnica Federico Santa María, Casilla 110V, Valparaíso, Chile
[3] Departamento de Física, Universidad de Murcia, Apartado 4021, 30080 Murcia, Spain
[4] Department of Engineering Physics and Mechanics, Kyoto University, Kyoto 606-8501, Japan
[5] Instituto Balseiro, Centro Atómico Bariloche, RA-8400 Bariloche, Argentina

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We have measured and calculated the energy distribution at zero exit angle of the proton fragments resulting from the dissociation of fast H_2^+ molecules when traversing carbon foils of different thicknesses. We discuss the role played by several processes that take place during the motion of the projectiles through the foil, in particular by the electron capture and loss processes, in order to explain the structure of the proton energy distribution.

1 Introduction The interaction of fast projectiles with solids provides a good tool to study characteristic properties of the target or the projectile. In particular, if the projectile is a molecular ion, during the interaction with the solid some characteristic effects appear associated with the closeness of the molecular constituents [1]. When a fast H_2^+ molecule penetrates into a solid it loses its binding electron in the first atomic layers [2], resulting in two protons in correlated motion. The interaction of each proton with the electrons and nuclei of the solid is influenced by the behaviour of its partner. Many consequences of this "vicinage effect" have been discussed both experimentally and theoretically (see Ref. [1] for a complete list of references) but there are still several phenomena that are worth to study. In particular, it has been established that when a H_2^+ molecular ion impinges on a solid the yield of neutral fragments is larger than the corresponding yield obtained with H^+ beams [3, 4]. Moreover, the analysis of the energy spectra of the fragments dissociated from the H_2^+ ions could give information about the charge state fractions of the hydrogen ions inside the target [5, 6].

In this work we present measurements and simulations of the energy distribution, at zero exit angle, of the proton fragments dissociated from fast H_2^+ molecular ions incident on carbon foils of different thicknesses. The projectile initial energy is 0.5 MeV/proton and the dwell time ranges from $\sim$1 to $\sim$6 fs. The aim of this paper is to analyze how the processes of electron capture and loss by the fragments dissociated from the H_2^+ molecular ions affect their energy distribution; moreover, the appearance of a small peak in the central region of the energy distribution and its dependence with the foil thickness can be understood in terms of these charge exchange processes.

[*] Corresponding author: e-mail: ias@ua.es, Phone: +34 96 590 95 78, Fax: +34 96 590 97 26

2 Experimental setup The experimental apparatus we have used was described previously [7], and so we will mention here only the specific characteristics of this experiment relevant to our subsequent discussion. A beam of 0.5 MeV/proton H_2^+ molecular ions from the 4-MV Van de Graaff accelerator of the Kyoto University was collimated by a series of apertures and introduced into a UHV scattering chamber (base pressure 4×10^{-10} Torr). The beam size and divergence were, respectively, less than 50 μm and 0.1 mrad on the target. The targets were self-supporting carbon foils with thicknesses in the range ∼2 to 10 μg/cm^2, which were estimated by measuring the energy losses of 0.5 MeV H^+ ions transmitted through the foils and comparing them with tabulated stopping power values [8]. The protons resulting from the dissociation of the H_2^+ ions upon transmission through the foil were collected and energy analyzed by a 90° sector magnetic spectrometer. The acceptance angle of the spectrometer was 0.02 μsr and its energy resolution was 0.1%, including the energy spread of the incident beam.

3 Computer simulation In order to calculate the energy distribution of the protons dissociated from a H_2^+ molecule in a solid we have improved a previous computer code [9, 10] to simulate the trajectory of the H_2^+ molecular ion at the beginning of its travel through the solid and the trajectories of each fragment after dissociated from the molecular ion.

Before the dissociation of the H_2^+ molecular ion there are two main interactions that take place between the projectile and the target: the self-retarding force (due to the electronic excitations induced in the solid) and the elastic collisions with the target nuclei. After an average time of 0.23 fs [11] the H_2^+ molecular ion dissociates into two fragments that travel in a correlated motion, suffering the following interactions: (i) self-retarding force, (ii) wake force due to the electronic excitations induced in the target by the partner fragment, (iii) Coulomb repulsion with its partner (only when both fragments are charged), and (iv) elastic scattering with the target nuclei. Outside the material the exiting fragments only feel the Coulomb repulsion when both emerge as protons.

Our computer code combines the numerical integration of Newton equations to describe the dynamics of the projectiles motion, and a Monte Carlo approach [12] to account for the elastic scattering of the projectiles with the target nuclei; the effect of coherent scattering is negligible in this energy range [9]. The self-retarding force acting on the H_2^+ molecular ion and the dissociated fragments, the wake force and the energy loss straggling was calculated within the dielectric formalism framework [13, 14]; details of the calculation for the different type of projectiles (H_2^+, H^+, H^0) can be found in Refs. [15, 16]. The response of the stopping medium to the motion of fast particles is described by a linear combination of Mermin type energy loss functions for the outer-electrons excitations and a generalized oscillator strength in the hydrogenic approach for the inner-shell electrons [14, 17, 18].

We have also considered the possibility that each dissociated fragment can capture or lose electrons during its motion through the target; the H^- fraction is negligible in the energy range studied here [19] and so the fragments can only be H^+ and H^0 (i.e., the fractions Φ_0 and Φ_+ of atomic hydrogen and protons, respectively, satisfy $\Phi_0 + \Phi_+ = 1$). In order to implement in the computer code the process of charge exchange, in which electrons are captured or lost by the fragments, at each timestep of the finite difference algorithm we use a Monte Carlo procedure to draw the fragment charge state (neutral or proton) according to a prescribed equilibrium neutral charge fraction Φ_0. When a fragment is in the neutral state it does not experience the Coulomb repulsion with its partner, but it feels a self-retarding force that is different from the one felt by a proton, therefore the wake force has been incorporated into our computer code considering the possible combinations when the fragments are both protons, both neutrals, or a proton and a neutral. When the fragments reach the rear surface of the foil they continue through the vacuum, until they reach the detector, without changing their final charge state and feeling a Coulomb repulsion only when both fragments are charged. We have "collected" only the energy spectrum of those protons whose exit angle was less than the spectrometer acceptance angle around the forward direction. These energy distributions where convoluted with a Gaussian whose standard deviation depends on the energy resolution of the spectrometer and the foil roughness coefficient, which are 0.1% and 10%, respectively. To obtain a good statistics in our results we need to simulate up to 3×10^6 histories.

4 Results In order to check the dependence of our simulations with the neutral fraction Φ_0, Fig. 1 depicts the histograms (for the cases $\Phi_0 = 0$, 0.005 and 0.01) of the energy distribution of protons dissociated from a 0.5 MeV/proton H_2^+ beam and collected by the detector after they leave at zero angle a 2.1 μg/cm^2-thick amorphous carbon foil. The two large peaks in the energy spectra are due to the Coulomb explosion (mainly at the exit of the foil), and their asymmetry in size is understood in terms of the wake force, which tends to align the trailing fragment behind the leading one [5, 9, 10]. When $\Phi_0 = 0$ (i.e., all fragments travelling inside the target are protons), the small amount of protons that appear between these larger peaks is produced mainly by the nuclear scattering with the target nuclei, which in part inhibits the alignment tendency of the wake force. However we can see how a central peak develops as the fraction Φ_0 of hydrogen atoms increases, because now the fragments do not repel each other in a nonnegligible part of the pathlenght through the solid, and mainly because then there is a growing number of protons that do not experience a Coulomb repulsion at the exit of the foil. Therefore, electron capture and loss by the fragments clearly affects the detailed shape of the exiting proton energy distribution. Comparing the simulations shown in Fig. 1 for different neutral fractions of dissociated fragments with the corresponding experimental data (represented by symbols), we have adopted the approximated value $\Phi_0 = 0.005$ as the one to do the calculations for all the foil thicknesses employed in the experiments. This value of Φ_0 is larger than the one found for protons in amorphous carbon at this energy ($\Phi_0 \sim 0.0025$ [19]), which agrees with the reported fact [3, 4] that the neutralization of molecular fragments is enhanced in comparison with the case of isolated protons. The difference between the neutral fraction resulting from molecular or ionic beams can be attributed to the electron capture enhancement due to the proximity of molecular fragments [20, 21].

In Fig. 2 we show the energy distribution of the proton fragments, resulting from the dissociation of 0.5 MeV/proton H_2^+ molecular ions, which reach the detector after traversing carbon foils of thicknesses in the range ∼2 to 10 μg/cm^2; all the energy distributions are normalized to have unit area. Again, the energy distributions present two large peaks corresponding to those fragments that have lost or gained energy, respectively, with respect to the initial energy, due to the Coulomb explosion process that tend to accelerate the leading protons and decelerate the trailing ones. The position of the peaks is not symmetric around the incident energy because each fragment loses energy due to electronic excitations induced in the solid by itself and by its partner; the trailing peak is higher due to the alignment tendency of the wake force. For thicker foil thicknesses the region between the peaks becomes more filled with particles due to the major importance of the multiple scattering at larger thicknesses, which broadens the angular distributions [9, 10]; the central peak of the energy distribution is more noticeable at small thicknesses because at large thicknesses it is masked by the latter effect. The ratio between both contributions (charge-exchange and elastic scattering) to the height of

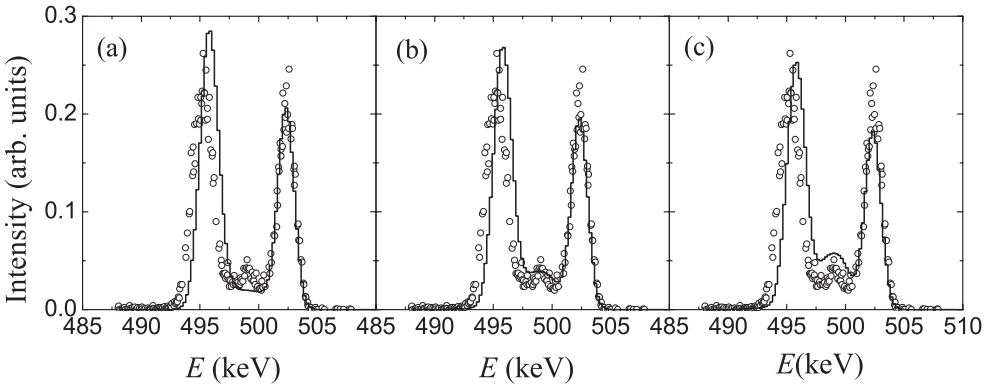

Fig. 1 Energy distribution of the protons dissociated from a 0.5 MeV/proton H_2^+ molecular ion, exciting at zero angle after traversing a 2.1 μg/cm^2-thick amorphous carbon foil. The circles correspond to the experimental data while the histograms represent the results of the simulation with three different values of the neutral fraction Φ_0: (a) 0, (b) 0.005 and (c) 0.01.

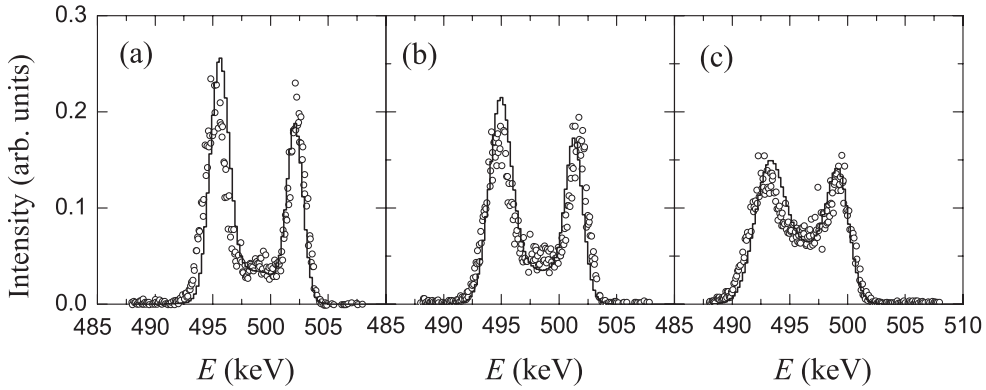

Fig. 2 Same as Fig. 1, for the foil thicknesses: (a) 2.6 μg/cm^2, (b) 4.35 μg/cm^2 and (c) 9.3 μg/cm^2. The simulations were done using $\Phi_0 = 0.005$.

the central peak is approximately 1, 0.5, 0.1 and 0, respectively, for the foil thicknesses 2.1, 2.6, 4.35 and 9.3 μg/cm^2. This central peak (which dissapears as we move from the thinner to the thicker foils) is due to those protons which have not undergone the Coulomb explosion process at the exit of the foil because their partner captured an electron just before leaving the target and emerged as a neutral particle.

The slightly smaller separation of the peaks in the simulations, as compared to the experimental data, can be explained by the fact that the experimental initial internuclear distribution of the H_2^+ molecular ion can be shrinked [22] in comparison to the theoretically calculated by us using the Franck–Condon principle; this would provide larger initial potential energies that lead to a larger separation of the Coulomb peaks. On the other hand, the expected asymmetry in the peak heights [23, 24] for the thinner foils does not appear experimentally possibly due to a problem with the energy calibration of the detector.

5 Conclusions We have presented experimental data and computer simulations of the proton energy distribution resulting from dissociated 0.5 MeV/proton H_2^+ in thin carbon foils of several thicknesses. The big peaks in the distribution are due to the Coulomb explosion process, whereas the small central peak appearing at the thinner foils is a consequence of the electron capture and loss processes, which frustrates the Coulomb explosion in a considerable part of the projectile pathlenght, mainly after exiting the rear surface of the target. The neutral fraction value that fits better to the experimental energy distributions is $\Phi_0 = 0.005$, this value is higher than the neutral fraction found experimentally for atomic hydrogen [19]. The simulations reproduce reasonably well the main features of the experimental data, confirming that the electronic capture and loss by the fragments is important to understand the shape of the dissociated-proton energy spectra.

The comparison of experiments and simulations as those reported in this work could be used to give a rough estimation for the charge fraction of a given experimental situation by adjusting this value to fit the central peak in the energy distribution of the protons dissociated from fast H_2^+ molecular ions.

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